# FINAL PUBLISHABLE REPORT

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| 1. PTB, Germany | 1. AGES, Austria | 1. IDEAS, Hungary |
| 2. BFKH, Hungary | 2. CLOR, Poland | |
| 3. CMI, Czech Republic | 3. INESC TEC, Portugal | |
| 4. ENEA, Italy | 4. JRC, European Commission | |
| 5. IFIN-HH, Romania | 5. LUND, Sweden | |
| 6. NPL, United Kingdom | 6. SUJCHBO, Czech Republic | |
| 7. VINS, Serbia | 7. UC, Spain | |
| | 8. UoB, United Kingdom | |
| | 9. UPC, Spain | |
| | 10. UVSQ, France | |

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# 1 Overview

Radon gas is the largest source of public exposure to naturally occurring radioactivity, and concentration maps based on atmospheric measurements aid developers to comply with EU Basic Safety Standard Regulations (EU-BSS). Radon can also be used as a tracer to evaluate dispersal models important for identifying successful greenhouse gas (GHG) mitigation strategies. To increase the accuracy of both radiation protection measurements and those used for GHG modelling, traceability to SI units for radon release rates from soil, its concentration in the atmosphere and validated models for its dispersal are needed. This project has provided the necessary measurement infrastructure and used the data generated to apply the Radon Tracer Method (RTM) which is important for GHG emission estimates that support national reporting under the Paris Agreement on climate change.

# 2 Need

An overlapping need exists between the climate research and radiation protection communities for improved traceable low-level outdoor radon measurements, combining the challenges of collating and modelling large datasets, with setting up new radiation protection services. Compared to the large spatiotemporal heterogeneity of GHG fluxes, radon is emitted almost homogeneously over ice-free land and has a negligible flux from oceans. Radon flux relates to the transfer process of radon activity from soil to the atmosphere per square metre and second, whilst radon activity concentration is the amount of activity of radon in the atmosphere per cubic metre. Atmospheric measurements of radon activity concentrations can be used for the assessment and improvement of atmospheric transport models (ATM). However, traceability to the environmental level did not currently exist for measurements of radon fluxes and atmospheric radon activity concentrations. Therefore, significant improvements in such measurements were needed. Climatic Atmospheric Monitoring Networks (AMN) like the European Integrated Carbon Observation System (ICOS), are infrastructures that operate GHG monitoring stations and include atmospheric radon monitors in their stations. The radon data produced from such networks can be used to improve transport modelling and the estimation of GHG emissions based on the RTM, which uses the correlation between GHG and radon concentrations. However, this radon data needed significant improvement in terms of the accuracy of both radon flux measurements and environmental radon activity concentrations in the range 1 Bq m$^{-3}$ to 100 Bq m$^{-3}$ to be able to provide robust data for use in the RTM. Similarly, for radiation monitoring, all European countries have installed networks of automatic radiation dose and airborne contamination monitoring stations and report the information gathered to the European Radiological Data Exchange Platform (EURDEP), thus supporting EU member states and the EURATOM treaty. Currently, monitoring information on dose rates is collected from automatic surveillance systems in 39 countries, however, urgently needed data on outdoor radon activity concentrations is not yet collected due to a lack of ability to measure accurately at the low levels encountered in the environment. Furthermore, accurately detecting contamination from nuclear emergencies relies on rejecting false positive results based on radon washed from the atmosphere by rain. Therefore, improving contamination detection requires greater accuracy in determining environmental radon concentrations and their movement in the atmosphere. This project has provided new traceability chains from lab to field for the radon activity concentration and established a traceability chain for radon flux for the first time. This has triggered lots of interest worldwide from stakeholders wishing to include these approaches for quality assurance into their climate observation or radiation protection capabilities.

# 3 Objectives

The overall aim of this project is the development of metrological capacity (reference monitors, transfer standards and robust methodology) to measure low levels of radon in the environment, which can be used to determine emission reduction strategies of GHG and improve radiation protection of the general public.

The specific objectives are:

1. To develop traceable methods for the measurement of outdoor low-level radon activity concentration in the range of 1 Bq m$^{-3}$ to 100 Bq m$^{-3}$, with uncertainties of 10 % for k=1, to be used in climate monitoring and radiation protection networks. These methods include two new traceable Rn$^{-222}$ emanation sources below 100 Bq m$^{-3}$, a transfer instrument calibrated with these new sources to assure the traceability of the transfer instrument and a calibration procedure suitable to enable a traceable calibration of environmental atmospheric radon measurement systems in the field.

2. To develop the capability for traceable radon flux measurements in the field, based on the development of a radon exhalation reference system “exhalation bed” and a transfer standard (TS). To use this capability to harmonise existing radon flux instruments/methods by intercomparison campaigns. To develop a first standard protocol for the application of the radon tracer method (RTM) to enable retrieval of greenhouse gas fluxes at atmospheric climate gas monitoring stations and to use radon flux data for the identification of Radon Priority Areas (RPA).
3. To validate current radon flux models and inventories by the new traceable measurements of radon activity concentration and radon flux. To support the validation with dosimetric and spectrometric data from the radiological early warning networks in Europe. To improve process-based radon flux maps that will be available for use in the RTM, atmospheric dispersion modelling, and radiation protection.
4. To provide easy to use dynamic radon and radon flux maps for climate change research and radiation protection in line with Council Directive 2013/59/EURATOM, including their use to identify RPA and radon wash-out peaks.
5. To facilitate the take up of the technology and measurement infrastructure developed in the project by the measurement supply chain (NMIs, calibration laboratories), standards developing organisations (e.g. IEC, ISO) and end users in greenhouse gas monitoring and European radiological early warning networks.

# 4 Results

**Objective 1:** Traceable measurements of outdoor radon activity concentrations

**Lead:** PTB

**Detailed description of project developments against objective 1:**

The first part of the new traceability chain given in [1,2] was performed by NPL with support from PTB, UVSQ, UPC and IDEAS. It encompasses a literature study of currently available $^{222}$Rn emanation sources for calibration of instruments capable of measuring $^{222}$Rn activity concentrations below 100 Bq·m$^{-3}$. The intention was to identify existing low-level $^{222}$Rn emanation sources, their potential use, and their most beneficial characteristics, so that the latter could be implemented into the low-level $^{222}$Rn emanation sources, that were to be developed in the next activity.

The most reported $^{222}$Rn sources for calibration of $^{222}$Rn instruments are solid Pylon sources and standard reference material provided by the National Institute of Standards and Technology (NIST) (references [3]–[9]). None of the sources found in the literature review were suitable for the traceable calibration of instruments at radon activity concentrations below 100 Bq·m$^{-3}$. The radon monitors on the other hand were more promising. Most are capable of measuring activity concentrations below 100 Bq·m$^{-3}$ as specified by the manufacturer and confirmed in practical use. From the literature review the AlphaGUARD $^{222}$Rn detector had already been reported to be used as a secondary standard but with the restriction of low time resolution or higher activity concentrations.

Both the existing sources and existing measurement instruments showed applicability in a wide range of temperatures, pressures and humidities typically found in the earth’s atmosphere. They can be classified into two groups: detectors operated in a temperature-controlled shelter (typically 15°C to 30°C) and those operated outside with a possible temperature range of -20°C to 60°C. In both groups the pressure ranges between 620 hPa and 1050 hPa and the humidity between 0 % and 100 %. The two new sources should be capable of providing reliable calibration under similar conditions. Special care must be taken with regard to humidity as it was found that the emanation factor of $^{222}$Rn is highly dependent on it. The more the humidity rises the more $^{222}$Rn tends to be released from the source into the measurement chamber. The results were published in references [10] and [11].

The next step was to implement these results to design and develop two new traceable low-level $^{222}$Rn emanation sources for delivering radon activity concentrations below 100 Bq·m$^{-3}$. They were supposed to be suitable as calibration standards in the field or in Rn calibration chambers and capable of producing stable reference atmospheres. Both, PTB and CMI, developed one of the sources with support from SÚJCHBO. They will be described in the following.

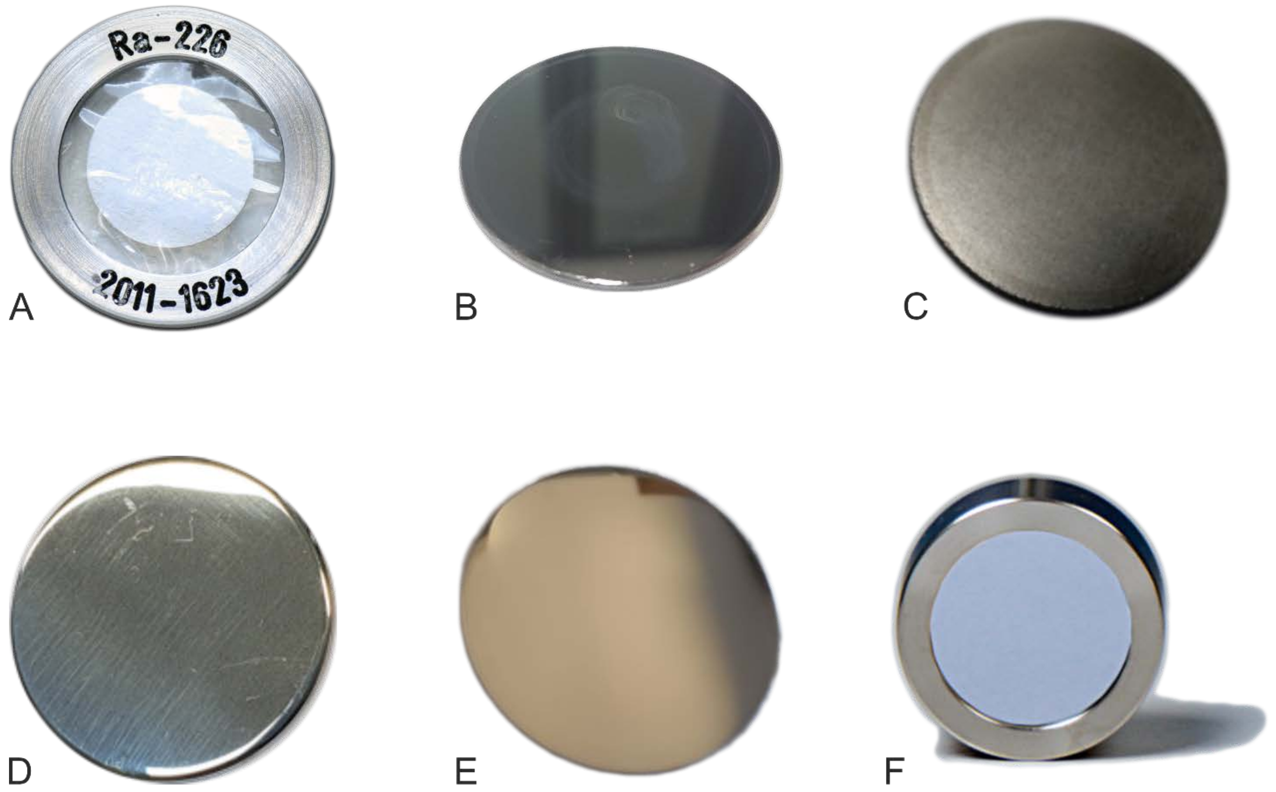

Figure 1: Pictures showing the development of the different $^{222}$Rn emanation soures at PTB. Source F shows the newsest combination of Source and detector (IRSD).

At PTB a new technological approach was chosen: an integrated source-detection system operated in real time monitoring mode, referred to as Integrated Radon Source Detector (IRSD) (see also reference [12] and Fig.1). For the first time, this novel combination of source and detector developed at PTB was used. For this purpose, an ion-implanted silicon semiconductor detector was coated in a defined manner with radium chloride ($^{226}$RaCl$_2$), by means of thermal vapor deposition directly onto the dead layer of the detector. Thus, the detector itself is the source of the radon. At the same time, it is the spectrometric detector for the resulting alpha radiation. The results were published in references [12], [13] and [14].

Both, the absolute activity of $^{226}$Ra and the loss of $^{222}$Rn, can be determined directly by analysis of the α-spectrum of the IRSD. This yields the absolute activity of $^{222}$Rn emanating from the integrated source-detection system. Therefore, the source developed at PTB is a low-level $^{222}$Rn emanation source that can be directly implemented to create a reference atmosphere. The procedure is described and published in reference [14].

CMI on the other hand implemented a different approach with a $^{222}$Rn source at higher concentration, which is diluted with air before $^{222}$Rn gas is emitted into the reference atmosphere (see Fig. 2). In the 1950s, F. Kysela from the Institute for Research, Production and Utilization of Radioisotopes (ÚVVVR) in Prague created a primary set of 13 $^{226}$Ra sources by filling RaBr$_2$ into Thuringian glass tubes with a diameter of about 5 mm and wall thickness of 0.27 mm. Four of these emitters were compared to the standards of Prof. Otto Hönigschmid, deposited in Vienna, in 1957. The traceability was performed by Prof. Dr Berta Karlik from the Institut für Radiumforschung und Kernphysik. The remaining nine sources were compared to these four sources. The low-level radon emanation source was created from one of the nine sources. An emulsion of salts of fatty acids in silicone rubber was formed from the weighed standard solution. The emulsion was allowed to polymerize in a steel tray with the following dimensions: 70 mm × 30 mm. The activity of the standard was determined by the weight of the $^{226}$Ra-solution, the weight of the resulting emulsion and the losses (< 0.1 %). The whole process was controlled by weighing and gamma spectrometry on an HPGe detector. The 185 keV gamma-ray emission intensity was measured with the use of the standard solution, which confirmed excellent conformity with the tabulated value. The source was constructed as a stainless-steel cylindrical case, supplied on the ends with ball valves and two aerosol filters connected on the output aperture of the valves. The steel tray with $^{226}$Ra was placed in the middle of this cylindrical case, and radon was released from this thin layer.

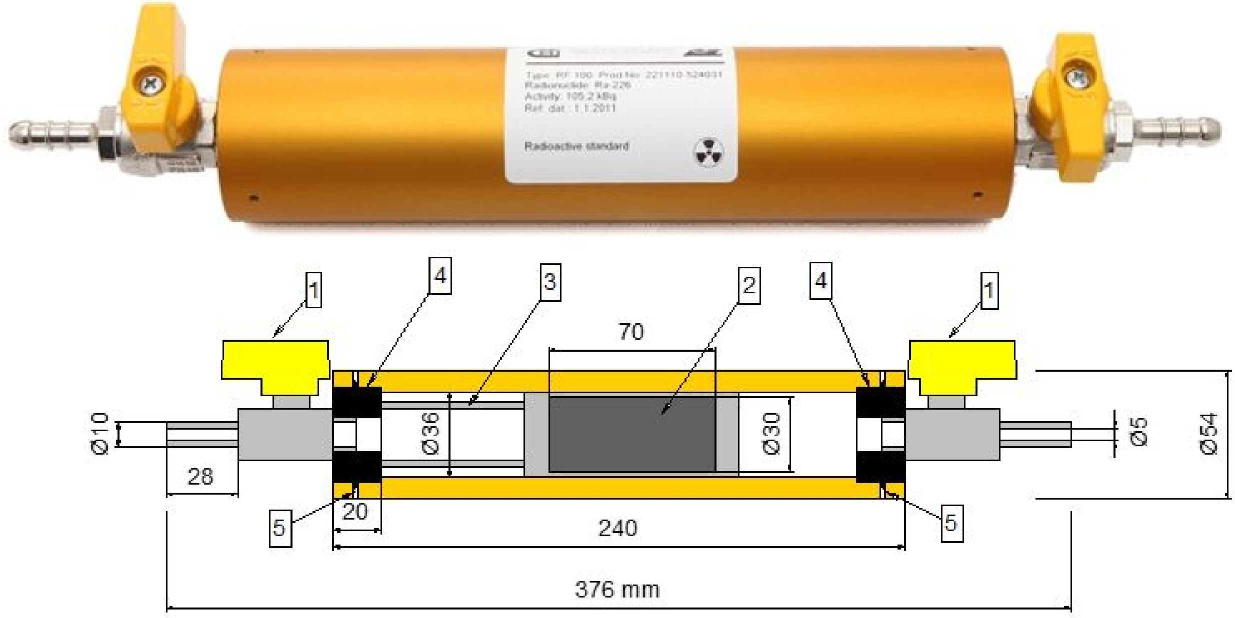

Figure 2: Picture and schematic drawing of the $^{222}$Rn emanation source developed at CMI.

Successful application of the sources to create reference atmospheres required them to be thoroughly characterized. The methodology and the calibration method were reviewed by BFHK, ENEA, CMI, IDEAS, SUJCHBO and commented on by UPC, ANSTO, CLOR and IFIN-HH. The protocol for the comparison of the sources has been developed by SUJCHBO, CMI and PTB mainly and agreed by all partners.

Both sources were studied at PTB and SÚJCHBO.

To ensure comparability a comparison protocol was developed beforehand. Since the literature review recommended the AlphaGUARD radon detector, radon reference instruments (RRIs) of this type were chosen for the measurements at SÚJCHBO and PTB. The implemented setups and resulting calibration factors will be illustrated in the subsequent outline.

At PTB the RRI #1 (AlphaGUARD 626) was placed inside a 50 L closed reference volume and operated in diffusion mode. The IRSD #1 was connected to the reference volume through a standard vacuum KF40 flange T-piece, whereas the CMI-source was placed directly inside the reference volume. For further comparison a second setup (setup #2) was made. Another radon reference instrument (RRI #2, an AlphaGUARD 1950) and another IRSD (IRSD #2) were placed inside a 500 L closed reference volume. The rest of setup #2 was analogue to setup #1 as described above. Comparison of both sources was carried out based on the derived values of the RRI calibration factors, $k_1$ and $k_2$ for setup #1 and setup #2, respectively, with respect to each sources certified activity and emanation rate. In the ideal the case calibration factors determined for the respective RRI by implementation of the differing emanation sources would be identical, as both sources are meant to be suitable as calibration standards and should yield the same calibration factor for the same RRI. The activity and emanation factor of the CMI-source were taken from the issued calibration certificate of CMI (see reference [15]), whereas the PTB development, the IRSD, allows for retrospective, data-driven computation of the volumetric activity concentration. Although both sources were thoroughly characterized the measurements result in differing calibration factors for all RRI. It is remarkable that the calibration factors for the respective RRIs determined by implementing the IRSD are slightly lower than those determined by implementing the CMI source in all setups. Since the statistical uncertainty of setup #1 is far below 1 %, it is unlikely for this to be the cause of the differing calibration factors. On the other hand, the uncertainty of the fit is on the order of 1% and therefore, a possible explanation. For the measurements at SÚJCHBO a slightly different approach was chosen, which will be described in the following section. The measurements were made under laboratory and under field conditions. The laboratory conditions are described in reference [16]. A newly developed piece of equipment is now part of the Czech primary radon measurement device situated in SÚJCHBO, v.v.i. Kamenna (Central Bohemia). In particular, the equipment consists of an airtight low-level radon chamber (LLRCH), a humidifier, the respective $^{222}$Rn source, a mass flow controller of the Bronkhorst® EL-Flow type (Bethlehem—PA, USA), an aerosol filter and an air pressure vessel as the source of radon-free air. For better comparison a similar setup was chosen under field conditions. The respective source was connected to an AlphaGUARD (RRI #1) and measured in flow-through mode. In addition, a second AlphaGUARD (RRI #2) was implemented for the purpose of background measurements. The measurement procedure consisted of three phases: During the first phase, both RRI were used to measure the air flow without a Rn source. As a result, both should ideally measure the same (outdoor) Rn activity concentration. During the second phase, the setup of RRI #2 remained unchanged, but RRI #1 was connected to the Rn source. In the third Phase. The source was disconnected from RRI #1 such that both RRI were then NOT connected to the Rn source. Based on a comparison of the subsequent measurements of RRI #1 and RRI #2, it was possible to check the outdoor radon activity concentration as measured in diffusion mode. The measurements proved that the sources could provide stable reference atmospheres (indoors and outdoors) below 100 Bq·m$^{-3}$. The two radon emanation sources developed in the framework of the project were successfully and thoroughly characterized regarding their suitability as low-level calibration standards.

The calibration factor $k$ was chosen as the comparison parameter. At PTB and SÚJCHBO the IRSD resulted in a slightly smaller $k$ for the respective RRI than the $k$ determined using the CMI source (of the order of 2 %). Furthermore, all calibration factors were within the aspired goal of an uncertainty of smaller than 10 % for $k$ = 1. The comparison was finished successfully.

Since the quality of the two new $^{222}$Rn emanation sources had been confirmed, they could be used to produce a temporally consistent $^{222}$Rn atmosphere below 100 Bq·m$^{-3}$ in a calibration chamber. This was a necessary requirement to reach the overall objective: To develop a methodology for the traceable calibration of atmospheric radon monitors including the means to implement it. Four different procedures to provide traceability to the SI were presented:

Procedure 1: A primary method based on a reference activity concentration realised by a primary radon gas standard and a calibration volume (both values are traceable to national standards). For this method the absolute activity is measured with an α-particle spectrometer in a reference volume with a radon activity standard. As a result, the activity is already known before the measurement is made. The expected and the measured value are linked by the calibration factor (which is ideally 1, meaning both measurements are identical). The measurement device investigated in such a way becomes a secondary standard.

Procedure 2: A secondary method based on calibration via a reference monitor enclosed in the same atmosphere as the system under test. This procedure is essentially based on procedure 1 as it implements a secondary standard (calibrated for instance by procedure 1) and uses it to determine the reference activity for the calibration factor.

Procedure 3: A primary/secondary calibration in a constant atmosphere based on a radium emanation source. This method being primary or secondary with respect to the components used. It is a long-term procedure (8 – 10 half-lives of $^{222}$Rn), as radioactive equilibrium is needed. It can be operated in a closed system or in an open system. The latter was used to create reference atmospheres based on the CMI-source, as its construction allows the time-stable radon activity concentration to be maintained at a precise level for several days in a radon chamber.

Procedure 4: A primary calibration in a non-constant atmosphere based on a radium emanation source in a closed volume. The system under test needs high sensitivity. This procedure is the only new procedure presented here and it is based on procedure 3. It is operated in a closed system implementing the IRSD from PTB, which allows for data-driven computation of the volumetric activity concentration. As a result, it is no longer necessary to wait for equilibrium to be reached, as the procedure is based on data measured during the build-up phase.

Four different reference atmospheres have been created at PTB. All of them were measured by the same instrument and yielded the same calibration factors for this instrument. They are therefore consistent and suitable to provide traceability to the SI. The presented results on calibration method for the creation of a temporally consistent $^{222}$Rn atmosphere below 100 Bq·m$^{-3}$ with uncertainties of 10 % ($k$ = 1) in a low-level radon calibration chamber with two newly developed radon sources and covers the objective “Traceable measurements of outdoor radon activity concentrations”. Both, the calibration method, and the new sources, were successfully developed and validated. Additionally, to the intention of the objective a full analysis of the characteristic limits was performed by analytical calculation as well as Monte Carlo simulation. Both approaches were applied for the decision threshold and the detection limit. This is considered a substantial additional benefit for the traceability chain and involved a lot of effort in the determination of the intrinsic background reading.

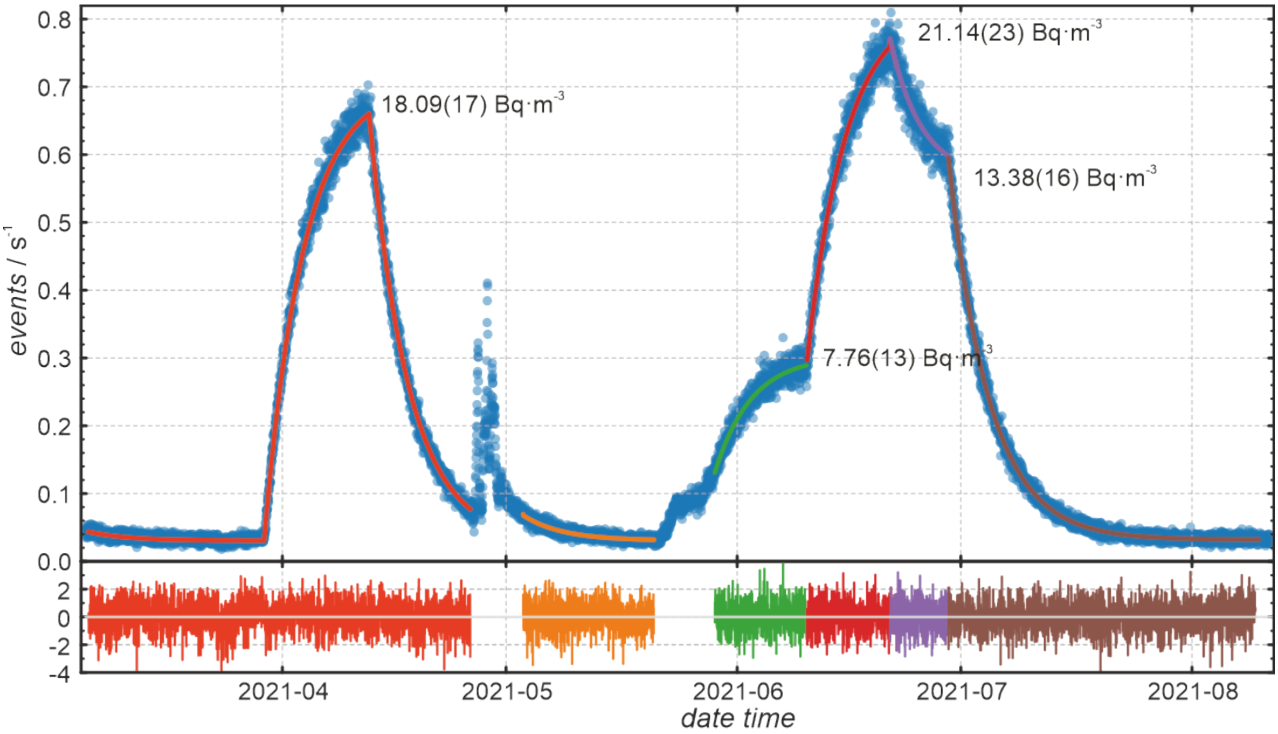

Figure 3: Calibration of the TS ANSTO 200 L performed at PTB. The upper graph shows the measured events per s of the detector (blue dots) and calculated model (colored lines). The lower graph shows the difference between model and measurements.

The achieved reference atmospheres are the base for the traceable calibration of the transfer standard: Originally only one transfer standard (TS) was planned. The success of the project was so significant, that two different types of TS could be developed instead: The ANSTO 200 L from ANSTO and the ARMON from UPC. First a literature review of currently available $^{222}$Rn monitors capable of measuring activity concentrations below 100 Bq m$^{-3}$ was performed. It was meant to become the foundation for a matrix of properties for the field application of these $^{222}$Rn monitors. Furthermore, it was used to identify the required parameters for a TS for the traceable calibration of atmospheric $^{222}$Rn monitors according to IEC 61577. In total, six $^{222}$Rn monitors were provided by UVSQ, ENEA, NPL and PTB. This determined whether any of the $^{222}$Rn monitors was suitable for use as a TS for the traceable calibration of atmospheric $^{222}$Rn monitors, and if so which of the $^{222}$Rn monitors is the best for use as a TS (see Fig. 3). As a next step, the property matrix was used to develop and build portable $^{222}$Rn monitors for measuring atmospheric $^{222}$Rn activity concentration below 100 Bq m$^{-3}$. A thorough characterization was performed, including determination of uncertainty and characteristic limits of the $^{222}$Rn monitors. A blank indication test to determine the lower limit of $^{222}$Rn activity concentrations, that can be reliably measured by the TS was performed at PTB.

The identification of the training needs of ICOS and other AMNS for $^{222}$Rn calibration and operation of $^{222}$Rn monitors at AMNS was added. For this purpose, a survey was designed for operators of AMNS to establish existing practices for the traceable calibration of $^{222}$Rn instruments used to determine atmospheric $^{222}$Rn activity concentrations. The survey also investigated the local ranges of environmental parameters: Based on these matrices PTB, NPL, UoB, ENEA and UVSQ have provided suitably identified radon monitors to be used at their stations for continuous measurements of radon activity concentration.

On this basis, a summary of possible options for traceability chains at AMNS was made and used to propose an optimized traceability chain for environmental atmospheric $^{222}$Rn activity concentration monitoring measurement systems suitable for deployment at AMNS. Two TS fulfilled the need: The first instrument, ANSTO 200 L, that was developed within this objective is a novel, portable (200 L) direct (radon gas) dual-flow-loop two-filter radon monitor. This instrument has a 30-minute temporal resolution and a sensitivity of (0.0385 ± 0.0020) s$^{-1}$ Bq$^{-1}$ m$^{3}$, for the first time, is traceable to the new sources. While it only measures $^{222}$Rn, this instrument is fully remotely controllable, can fit within a 19” instrument rack, has low power requirements (~100 W at 240 VAC), is suitable for low-maintenance long-term indoor or outdoor operation, records internal environmental parameters for STP and water vapour correction of radon concentrations, and has the capability to perform calibrations/instrumental background checks automatically in situ. The second instrument, ARMON, that was developed in this objective is from the Universitat Politècnica de Catalunya (UPC). Progress has been made to develop the detection, acquisition and drying sample modules of a new pre-prototype instrument. The new modules were based on the previous model of the Atmospheric Radon MONitor (ARMON) used for the measurement of atmospheric radon and thoron ($^{220}$Rn) concentrations. Laboratory experiments were performed at the UPC radon chamber to test the PIPS detector, the detection volume, the electronics, the high voltage and the drying system components. In addition, a theoretical study of the electrostatic field generated within the detection volume was performed to improve its geometry and maximize collection of the $^{218}$Po and $^{216}$Po on the detector surface. Finally, a GUI has been created to remotely control the different modules and to visualize the results in real time. Results indicate a sensitivity of this pre-prototype of about 0.006 $s^{-1}$ $Bq^{-1}$ $m^3$ for radon concentration, with a detection volume of only 20 L.

To compare the results of atmospheric $^{222}$Rn measurements between the developed TS instruments a long-term comparison (6 months) with an existing operational $^{222}$Rn instrument used to measure environmental $^{222}$Rn activity concentrations at UVSQ was performed. As further quality assurance, a one-month stability comparison of the TS was carried out at two different AMNS (UVSQ and PTB, which was accepted as a replacement for an AMNS). The aim was to prove consistency of the conventional true value of the TS and that of the $^{222}$Rn activity concentration monitor of each AMNS. To determine the integral $^{222}$Rn exposure during the comparisons, five solid state nuclear track detectors (SSNTD) traceable to the national $^{222}$Rn chamber of IFIN-HH were implemented. Based on the results, an overall uncertainty budget for traceable atmospheric $^{222}$Rn activity concentrations measured at ICOS stations and other AMNS and their ability to achieve an hourly uncertainty below 15 % for $k$=1 was made. In a final step, a calibration procedure for the traceable measurement of atmospheric $^{222}$Rn activity concentration in the field with a specific focus on the needs of ICOS and other AMNS and including how to best establish a traceability chain, either directly by source or by using a transfer instrument was developed.

In conclusion, the objective was fully met, and the planed work was extended being so targeting and successful. The results overachieved the expectations: Two new source types are available, a completely new type of instrument, the IRSD was developed. The IRSD provided the ground for a new calibration method, which is faster and more accurate. By this, traceability to two new TS was generated and the traceability to the SI is available in field for the first time. All partners (PTB**,** BFKH, CMI, ENEA, IFIN-HH, NPL, CLOR, SUJCHBO, UoB, UPC, UVSQ, IDEAS) collaborated in the generation of these results.

**Objective 2:** Radon flux measurements

**Lead:** UPC

**Detailed description of project developments against objective 2:**

The radioactive noble gas radon ($^{222}$Rn) is known to contribute over half of the total public exposure to radiation dose from natural sources. In addition, due to its short half-life (3.8 days) and its chemical inertness, this gas is being widely used as an environmental tracer both for atmospheric and geophysical processes. Particularly, climate scientists are using co-located measurements of atmospheric radon and GHG concentrations to apply the RTM for the purpose of retrieving GHG emission estimates [17,18]. Each of the above applications requires information, with the lowest achievable uncertainty, regarding the amount of radon exhaled per square meter of soil per unit time over an area of interest to quantify its emission into the atmosphere. This quantity is referred to as the radon flux, $F$ (or radon exhalation rate, $E$) and its SI measurement is in Bq $m^{-2}$ $s^{-1}$ (or mBq $m^{-2}$ $h^{-1}$). After its formation by $^{226}$Ra decay, $^{222}$Rn escapes from soil pores to the atmosphere e.g. by diffusion. $^{222}$Rn exhalation rates depend strongly on the content of $^{226}$Ra in the soil and on the soil properties (porosity, tortuosity, soil humidity, etc). Therefore, the $^{238}$U content as well as the parameters influencing diffusive transport characteristics of the soil need to be known to properly estimate the variability of $^{222}$Rn exhalation rates [19-21]. In addition, the radon emanation factor has been observed to change with soil humidity (also known as water content of the soil) [22, 23]. Furthermore, numerous other interrelated factors affect the radon flux from a soil surface, including soil type, atmospheric pressure, rainfall (related to soil moisture), and soil temperature. However, complex dependencies between these factors makes it difficult to quantify the change in radon exhalation due to any one of these factors in particular (e.g., a precipitation event is often also associated with a drop in pressure and temperature).

Although the primary driving force of radon transport is diffusion due to the radon concentration gradient, advective processes may also occur. These processes are the result of pressure gradients, the effect of changing wind speeds, etc. Advective processes could therefore have an influence on radon flux measurements [24]. To date, most radon flux studies have been based on random sampling and temporal measurement data, which is not sufficient to clarify the relationship between radon flux and environmental factors. This is a contributing factor to why some studies reach contradictory conclusions about the influence of individual parameters on the radon flux. Long-term ongoing measurements are needed where radon flux measurements can be carried out together with observations of environmental parameters [25]. It is necessary to consider these requirements when a radon flux measurement system is selected [26]. There was a lack of a robust metrology for the measurement of radon flux from the soil surface. It was an aim of the objective, to overcome this. The objective aims to create the necessary infrastructure to enable high frequency in situ radon flux measurements from soils with a traceability chain to the SI.

In order to accomplish this goal, the following steps were undertaken.

i) a literature review was performed by UC on existing exhalation bed (EB) facilities and requirements,

ii) this review informed the design and construction of an exhalation bed facility

iii) a literature review of available radon flux monitoring systems and their main requirements was performed by UC, BFKH, UVSQ, LUND, ENEA, IFIN-HH, PTB, SÚJCHBO, IDEAS and collaborator ANSTO

iv) this review paved the way on the selection and improvement of possible radon flux transfer standard (TS) devices

v) the performance of the TS was characterized under laboratory conditions using the EB by UC;

vi) the combination of TS and EB were used to calibrate other radon flux monitoring systems and a calibration protocol was developed by UPC, UC, IFIN-HH and ENEA;

vii) the response of the TS and other available radon flux systems was compared at a reference site and guidelines were generated for in situ radon flux measurements [27].

The TS and EB facility enable the calibration of radon flux systems with a total calibration factor uncertainty of 6.4 % ($k$=1) for radon fluxes in the order of a thousand Bq $m^{-2}$ $h^{-1}$. Radon flux systems used for in situ radon flux measurements should fulfil the following specifications:

- A continuous radon monitor working in flow mode, with a low internal background, a temporal resolution not higher than 10 minutes, high precision, and, preferably, the ability to distinguish between radon and thoron contributions.
- An accumulation chamber that can be opened automatically at a set time interval, with a collar that can be correctly installed into the soil, with environmental sensors to monitor conditions inside and outside of the chamber as well as in the surface soil layer, painted in a reflective color to minimize solar heating of chamber air, with an effective height no bigger than 0.2 m to avoid low radon concentrations inside the chamber;
- A previous calibration under laboratory conditions using a TS and the EB, or being exposed directly in the field together with the TS for the transfer of the calibration/sensitivity factors;
- Application of the developed protocol when the monitor is used in field measurements to determine the maximum accumulation time to be applied for the linear fit method to be reliable;
- The use of a thoron delay volume in cases where the monitor is not capable of selective $^{222}$Rn measurement when $^{220}$Rn is also present in the air. However, the user should be mindful of the fact that this delay volume also delays the temporal evolution of radon concentration in the monitor.
- A system with an automatic arm to carry out radon flux measurement at different points could be also recommendable.

After all the preparations in the lab were finished the freshly calibrated radon flux systems were used in two intercomparison campaigns were conducted in northwestern Spain at low and at high radium content. The main goal of the experiments was to test the response of radon flux systems based on different monitors and different accumulation chambers to identify physical reasons for possible inconsistencies, particularly related to sampling and measurement techniques. The continuous radon flux monitoring capability was analyzed to harmonize the radon flux methods under field conditions. The participant institutions in this exercise were the UC, UPC, ENEA, IFIN-HH. Every institution participated with their own device, managed by themselves, except for UPC, which managed its own system and also an ANSTO Autoflux designed by ANSTO. All the systems involved in this study were made by a commercial continuous radon monitor (with or without pump) coupled with an accumulation chamber of different volume and shape, where the increase in the radon activity concentration over time was measured for calculating the radon flux.

Radon flux results obtained at the high radium content area by the different systems under study show significant differences among them. In addition, there is a large spread of radon flux values for some devices considering the individual measurements. The dispersion of the results may be explained by large and variable leaks observed in the systems and possible radon flux variability over time. Due to the short time of the campaign limited data was available. So, a final statistical analysis was not possible. The radon flux reference obtained by consensus may include a potential bias induced by each device's different number of measurements. However, the mean radon flux obtained in this area from the experimental observations was coherent with the value calculated using the so-called Karstens model developed from LUND after correcting the radium in soil content with the experimental one.

Radon flux results observed at the low radium content area provide interesting outcomes for common soils, usually presenting a similar average radium content. The results given by the different systems participating in the low radon flux area campaign are coherent among them and agree with the model prediction from Karstens at this site. The results seem to indicate that radon fluxes lower than 100 Bq $m^{-2}$ $h^{-1}$ should be measured with high sensitivity and high response time monitors to reduce the uncertainty of short-term measurements. Another option could be increasing the time considered in the linear fitting, which is possible in the case that leakages calculated by a 24 h experiment are small.

The results of the intercomparison campaigns indicate that the radon concentration evolution in the accumulation chambers of the systems is limited by the installation of the system, the radon monitors characteristics (diffusion or pump mode, integration time, sensitivity, etc.), and the features of the accumulation chamber (material, volume, tubes, etc.), although it was not studied in detail. The changes in the environmental conditions during the measurement also play a key role in the measurement and should be further investigated.

An overall agreement within the assigned uncertainties was reached within an interval of 20 %. This can be considered a first success, nevertheless the individual uncertainties were quite high, ranging from 15 % to 50 %, thus this agreement is a starting point for radon flux metrology, only. Overall, different radon flux systems were tested in the field under different radium content conditions. The work conducted contributes to the development of Good Practice Guide including a standard protocol for the measurement of radon flux and atmospheric activity concentration for application in the radon tracer method (RTM) for greenhouse gas (GHG) flux estimates and for its application to derive data for Radon Priority Areas (RPA) which was elaborated by UVSQ, NPL, UoB, UPC and LUND afterwards.

The Radon Tracer Method (RTM) has been used in many studies to evaluate the fluxes between atmosphere and ecosystems of trace gases such as $CO_2$, $CH_4$, $N_2O$ or $H_2$ originally by the University of Heidelberg. Historically, the RTM has been applied in one of two ways: either to investigate regional-scale fluxes on an event basis (where an event may span hours or days), or to investigate local-scale fluxes on a nocturnal basis. Here, as the aim is to propose an automated product, the focus is on the nocturnal accumulation RTM.

UVSQ with support from LUND, INESC TEC, NPL, JRC, UPC and collaborator ANSTO performed a review of the ICOS Atmospheric Monitoring Stations (AMNS) where radon activity concentration is currently measured, with the intention to choose the suitable AMNS for RTM evaluation. The evaluation consisted of the analysis of a set of radon activity concentration, GHG mixing ratio and meteorological data from the selected AMNS station measured over a broad range of atmospheric conditions. Supported was this work from the University of Heidelberg. The site selected in the first place for this task is the ICOS Saclay tower. Saclay (SAC) is located 30 km south-west of Paris, 48.7217°N, 2.142°E, 160 m asl.

The coding framework is written in python and is hosted on the ICOS Carbon Portal (CP) JupyterLab. By default, it uses the footprints already calculated without radon decay by the Lagrangian model STILT as configured on the CP (available for all ICOS sites and more for at least 2018 to 2020). The STILT footprints are available every 3 hours and cover the region 33°S-73°N, 15°W-35°E with a resolution of 1/12° by 1/8°, approx. 10 km x 10 km. The STILT model is forced with European Centre for Medium-Range Weather Forecast (ECMWF) Integrated Forecasting System (IFS) operational analysis. The radon exhalation maps used are either the InGOS one which is a climatology over 2006-2016 with one value per month or the two new maps developed in this objective (using either the reanalysed moisture data from ERA5-Land or GLDAS-Noah2.1) with a value per day and available from 2017 to May 2022. All maps can be downloaded at ICOS CP. The maps and the footprints use a different grid so when combined the radon exhalation map are regridded to the footprints. The site to study can be choosen from the list available on the CP. The RTM can be applied to several species when data are available ($CO_2$, $CH_4$, $N_2O$ and CO). Then either it extracts the data from the CP NRT hourly data or if access to the ICOS database with extraction rights is available, data with a smaller timestep can be extracted directly form the ICOS database.

By default, the code applies the RTM equation for the data between 21:00 to 06:00 UTC which is a suitable window for most sites in Europe, but this window can be easily modified to fit with other latitudes or longitudes. The length of the window can be modified as well for example to reproduce the tests from University of Heidelberg. No other criteria are applied but the correlation coefficient, the uncertainty on the linear regression, the number of data points and hours available for the calculation, the radon accumulation level and if the radon rise stopped before 08:00 UTC are recorded so the data can be filtered in a second step. For the sensitivity study, the possibility of using radon and greenhouse gas data from formatted files (csv) was added. Indeed, for the ANSTO detector, there is a measurement response time to consider, due to their design (a combined influence of their thoron delay volume, large measurement volume, and gross alpha counting approach). For optimal utilisation of radon measurements, a standardized protocol for data processing is required. This is not done yet in any ICOS radon data treatment chain. For this objective, a radon dataset derived from a preliminary standardized procedure was used, which is applicable to observations made by any similar (ANSTO made) radon monitoring system. The procedure to obtain the best estimate of atmospheric radon concentration (final product data) involves the traceability from objective one and the post-processing of radon data which includes the crucial deconvolution routine (step to correct for the instrument response time) as well as correction for standard temperature and pressure (STP). The possibility to use footprints from another model were added, too. For each model, it has to be tailored to it, depending on the grid size. The FLEXPART-WRF model version 3.3.2 [28], run at UPC, is used here. This model uses WRF meteorological files as inputs for its back trajectory calculations. This model was used with an output resolution of 0.05 degrees in order to fit with the new ERA-land and GLDAS-Noah2.1 radon maps. The back trajectories were calculated for a 24 h window time and assuming as footprint layer the 0 m - 100 m height. For the Saclay site, the spatial window used was [42.9 - 54.5] LAT and [-6 - 16.2] LON.

For the runs, the use of 3 different radon exhalation maps is available (called hereafter InGOS, traceRadon_ERA5, traceRadon_Noah), two models (CP-STILT, WRF-FLEXPART), two types of data (with and without the response time). Not all combinations are tested but all runs can go in pairs, with only one change from one to the other. Two months were chosen: February 2019 and August 2019 to observe the seasonal influence and as months with a good data coverage.

Different $^{222}$Rn fluxes for each night during the two months under study were used:

- constant radon flux value over the area of interest (52 Bq m$^{-2}$ h$^{-1}$);
- radon flux values obtained by available radon flux maps (InGOS, traceRadon_ERA5 and traceRadon_Noah) in the gridcell including the station. In the case of the InGOS map only a value for month was available where daily mean values are available for the two new traceRadon maps;
- radon fluxes values obtained coupling the previous radon flux maps with the ATM based footprints.

GHG fluxes within this study were calculated for every day during the months of February and August using, at least, two datapoints in the linear correlation between radon and $CO_2$. The linear fits calculated between nocturnal radon and $CO_2$ data at the Saclay stations were then filtered to retain only the meaningful events using the following criteria: $R^2>0.6$; error on the slope 50 %; radon increase over the night >1 Bq·m$^{-3}$. Results show winter fluxes are generally lower than summer ones as it was expected from the literature because of the lower water content in the soil during dry period. Daily radon fluxes based on GLDA-Noah reanalysis offer, for this station and periods of time, higher values than the ones calculated using ERA5-Land data.

Different runs were applied: Runs 1 and 2 were applied with the same input except that radon data from the 1500 L ANSTO monitor was used as calibrated detector output (not response time corrected) and as the best estimate (response time corrected) of the atmospheric radon concentration. This was done to study the influence of standardization on the efficiency of the RTM application. Runs 3 and 4 (yellow shaded cells in table 1) were carried out using footprints calculated with the same CP-STILT model configuration and the same atmospheric concentration radon and GHG data. In this case the radon flux maps traceRadon-ERA5 and traceRadon_Noah was used to evaluate how radon fluxes calculated using different soil moisture reanalysis data could influence the final results. Finally, run 5 (blue shaded grid in table 1) was executed with the same configuration of run 3 but using the FLEXPART-WRF based footprints which were calculated in the UPC cluster. Specifically daily fluxes vary between 12 Bq h$^{-1}$ m$^{-2}$ and 22 Bq h$^{-1}$ m$^{-2}$ for the run 3 and between 32 Bq h$^{-1}$ m$^{-2}$ and 40 Bq h$^{-1}$ m$^{-2}$ for run 4 while run 2 is at 20 Bq h$^{-1}$ m$^{-2}$ in February and between 48 Bq h$^{-1}$ m$^{-2}$ and 58 Bq h$^{-1}$ m$^{-2}$ for run 3 and between 67 Bq h$^{-1}$ m$^{-2}$ and 72 Bq h$^{-1}$ m$^{-2}$ for run 4 while run 2 is at 42 Bq h$^{-1}$ m$^{-2}$ in August. Radon flux results calculated using radon flux maps and ATM footprints show as expected a different variability but the range are in the same order of magnitude. In February, the fluxes vary between 19 Bq h$^{-1}$ m$^{-2}$ and 38 Bq h$^{-1}$ m$^{-2}$ for run2, 11 Bq h$^{-1}$ m$^{-2}$ and 42 Bq h$^{-1}$ m$^{-2}$ for run 3, 36 Bq h$^{-1}$ m$^{-2}$ and 71 Bq h$^{-1}$ m$^{-2}$ for run 4 and 15 Bq h$^{-1}$ m$^{-2}$ and 38 Bq h$^{-1}$ m$^{-2}$ for run 5. In August, the fluxes vary between 26 Bq h$^{-1}$ m$^{-2}$ and 64 Bq h$^{-1}$ m$^{-2}$ for run2, 31 Bq h$^{-1}$ m$^{-2}$ and 88 Bq h$^{-1}$ m$^{-2}$ for run 3, 44 Bq h$^{-1}$ m$^{-2}$ and 119 Bq h$^{-1}$ m$^{-2}$ for run 4 and 30 Bq h$^{-1}$ m$^{-2}$ and 115 Bq h$^{-1}$ m$^{-2}$ for run 5.

As can be expected, the variability on the radon fluxes is seen as well on the $CO_2$ fluxes. It is however interesting to notice that using the best estimate of atmospheric radon concentration, radon and GHG are more often seen as correlated and thus GHG flux can be calculated on more days. The standardized dataset allows to allocate the right sampling time for the radon measurement and thus when the two gases are influenced by the same air masses their correlation is better than when the data are not correcting and lagging behind. From this sensitivity test, it appears important to estimate the radon fluxes with at least the two different radon exhalation maps developed in the project to be able to estimate the range of uncertainties of the calculated fluxes. It is also important to use the standardized data when needed in order to obtain a more realistic correlation between GHG and radon. To apply the RTM, traceable obtained GHG and radon data has to be used STP correction on both gases have to be applied. For the ANSTO radon detectors, it is also necessary to apply a response time correction [29]. Then, the radon flux for the time window of the calculation has to be estimated. As the radon decay term can contribute to additional uncertainty, it should be directly calculated with the ATM models to obtain a value tailored to each situation. For RPAs, radon concentration measurement could be used in model inversion to validate the flux maps and therefore needs to be optimized as well. Within ICOS, the GHG data follow a standardized calibration procedure to ensure their quality. Uncertainty on the GHG measurement is very low compared to other terms.

Guidelines for the installation, calibration and operation of a radon and radon flux monitor have been developed to support their use, and the main recommendations for application of the RTM are:

- Using data from radon concentration measurements that is quality controlled.
- Using data from GHG concentration measurements that are quality controlled such as ICOS datasets.
- Using more than one radon flux exhalation map and if available more than one footprint model to estimate the radon flux uncertainty.
- If possible, comparing the radon flux with local measurements should be done within the mean footprint.
- Choose an adequate time window for the nocturnal accumulation gradient at your station: it can vary depending on the latitude/longitude of the station.
- Selecting data with a good correlation (i.e.$R^2$>0.6), and a significant radon concentration rise (i.e. above 1 Bq $m^{-3}$) to select meaningful events.
- Perform a sensitivity study as in [30] and here to evaluate the best criteria for an operational RTM calculation.
- Station sampling heights have to be taken into account; lower levels will allow to calculate local fluxes while higher levels may be decorrelated from the immediate surface during the night, being above the boundary layer height

In conclusion, the objective was fully met, traceability to the SI is established for radon flux measurements and the good practice for the application of the RTM for GHG flux estimates and for direct use in RPA is available. It has to be noted however, that the relatively large uncertainties in flux measurements due to practical problems on real soil, are still limiting the output from the RTM. Further research is necessary to open up the potential of the big data and machine learning in this field. All partners (UPC, PTB, BFKH, CMI, ENEA, IFIN-HH, NPL, CLOR, INESC TEC, JRC, LUND, SUJCHBO, UC, UoB, UVSQ, IDEAS) collaborated in the generation of these results.

**Objective 3:** Validation of radon flux models and inventories using radon flux and terrestrial data

**Lead:** LUND

**Detailed description of project developments against objective 3:**

To fulfil the need of validation of radon flux models and inventories using radon flux and terrestrial data the special sets of data had to be selected. This was done in close collaboration from LUND, JRC and supported by ENEA. To include new dosimetric and spectrometric data, with the aim to improve the model all partner of the consortium contributed.

Estimation of radon fluxes for the whole of Europe requires high-resolution data of the following parameters: Uranium content in the upper soil layers, the distribution of soil types and porosity in the unsaturated soil zone, and soil moisture and temperature as well as the water table depth. Soil uranium content from the European Atlas for Natural Radiation (EANR) [31] were provided as digital datasets by JRC on a 10 km x 10 km grid in Lambert azimuthal equal area projection covering Europe from the North Atlantic coast to »30° E. The map is based on uranium content in topsoil (0-25 cm depth) from the FOREGS [32] and GEMAS [33] soil sample measurement datasets.

The geographic coverage of soil uranium content information is expanded to cover Europe based on geological information available in the European lithological map [34-36] . A median uranium content was computed for the dominant lithological class in each of the overlapping grid cells of the two maps by LUND. The resulting relation was then used to extrapolate uranium content to the regions not covered by the map in EANR. Assigning a unique uranium content to each of the 30 lithological classes results in a relatively small spatial variability of the resulting extrapolated uranium content. Overall, this is a very indirect approach to estimate soil uranium content and hence associated with very large uncertainties, which will directly transfer into very large uncertainties in the final radon flux product. The extended uranium map can be updated as soon as more detailed information on the soil uranium content in countries not covered in EANR (e.g. Belarus, Romania, Russia, Ukraine, Turkey) becomes available. The parameterization of the radon source term requires the 226Ra activity concentration, which can be computed from the uranium content when assuming secular equilibrium between 238U and its daughter $^{226}$Ra. The conversion factor from uranium content to $^{238}$U activity concentration was taken from [37], i.e. 12.35 Bq kg$^{-1}$ per mg kg-1 uranium.

The physical properties of the soil were available as digital datasets through the European Soil Data Centre (ESDAC) at JRC. The European Soil database (ESDB v2.0) [38] covers Europe on a 1 km x 1 km grid in Lambert azimuthal equal area projection. Additional to soil type also derived data, like soil texture and bulk density, were made available through ESDAC. The regions outside the domain of ESDB (south-eastern part of the model domain) were complemented by information from the Global Soil Data Set for Earth System Modeling (GDSE); [39]. These soil properties are available in much higher resolution than the soil moisture reanalysis and can be used for application of the process-based radon flux model on the local scale, e.g. for comparison of the radon flux model with the radon flux measurement campaigns performed to fulfil objective two.

Soil moisture and temperature from two different state-of-the-art land-surface reanalysis datasets were pre-processed to be used as input for the process-based radon flux map. The soil variables are simulated by land surface models forced with meteorological fields from global reanalysis assimilating atmospheric measurements. Datasets from the following reanalysis products were downloaded for the European domain on their respective model grids and aggregated to monthly and daily means:

GLDAS-2.1 Noah Land Surface Model L4 forced by NOAA Global Data Assimilation System (GDAS) atmospheric reanalysis on 0.25°x0.25° grid [40,41].

ERA5-Land based on the H-TESSEL land surface model forced by the ERA5 reanalysis of the European Centre for Medium-Range Weather Forecasts (ECMWF) on 0.1°x0.1°grid [42,43].

Soil moisture and temperature are available for depth intervals of 0 cm - 10 cm , 10 cm - 40 cm, 40 cm -100 cm, 100 cm - 200 cm and 0 cm – 7 cm, 7 cm – 28 cm, 28 cm – 100 cm, 200 cm - 289 cm, respectively. Vertical means for the depth interval 0 cm - 40 cm were computed to be used in the radon flux model parameterizations.

Examples of monthly mean soil moisture (0 cm - 40 cm) reveal large differences between the two reanalysis datasets regarding geographic distribution. Area mean values for a region in central Europe show often higher soil moisture in the ERA5-Land reanalysis with a slightly larger seasonal amplitude in some years.

In order to apply the radon flux model in a consistent way, also the model-specific soil parameters, i.e. texture class and porosity per class, applied in the original land surface models are required. Porosities can regionally be very different in the two land surface models, e.g. in Scandinavia, which highlights the importance to use model-specific soil parameters together with soil moisture reanalysis. The water table depth dataset provided by [44] is based on observations of water table depth compiled from government archives and literature, together with a groundwater model to fill gaps and provide consistent patterns. Water table depth is only limiting the radon flux in areas where it is shallower than the diffusion depth, i.e. a few 10 cm to 1 m deep.

Radon fluxes for Europe were computed based on the two soil moisture reanalysis datasets, ERA5-Land and GLDAS-Noah 2.1, soil properties and uranium content according to the procedure described in Section 2. All input data sets were first remapped from their original grid resolutions and projections to a common regular latitude-longitude grid with a grid cell size of 0.05° x 0.05°. The resulting radon flux maps are provided on the same regular latitude-longitude grid as required for lower boundary conditions in atmospheric transport models. The monthly and daily radon flux maps for Europe are available online on the ICOS Carbon portal https://data.icos-cp.eu/portal/#%7B%22filterKeywords%22%3A%5B%22traceRadon%22%5D%7D where they can be downloaded and visualized.

In conclusion, the objective was fully met: Updated process-based radon flux maps for Europe are now available in high spatial (0.05°x 0.05°) and temporal (daily) resolution for the use in atmospheric transport model evaluation, in the RTM to estimate GHG emission trends, and in radiation protection applications. The validation was performed on the base of the campaigns. The timeline of a three-year projects limits the impact here. At four stations measurements have been performed but overlapping seasons would reveal much more information. Therefore, a fixed installation at the PTB reference site will be operated permanently. More detailed understanding of the transport processes in soil would be helpful to increase the benefit. All partners (LUND, PTB, ENEA, NPL, AGES, CLOR, INESC TEC, JRC, UoB, UPC, UVSQ) collaborated in the generation of these results

**Objective 4:** Radon and radon flux in maps for radiation protection issues

**Lead:** JRC/ENEA

In the radiation protection research area two applications of the use of radon outdoor concentration and radon flux data can be identified and the traceRadon project focused on them.

The first application regards the use of atmospheric radon concentration and radon flux to estimate the Geogenic Radon Potential (GRP) and to identify Radon Priority Areas (RPAs). The European Council directive 2013/59/Euratom [45], Article 103, Paragraph 3 states that Member States should identify areas where it is expected that annual average indoor radon concentration will exceed national reference level in a significant number of dwellings (EU, 2013). The reference level indicated in the BSS is 300 Bq·m$^{-3}$. These areas are often called Radon Priority Areas (RPA). The delineation of these areas will allow the planning and prioritization of reduction measures within the national action plans (which is another element of the Directive, to be established by EU Member States) and has implications on the required radon measurements in workplaces located in these areas. Further to legally binding requirements (workplaces), such a prioritization can also be useful for radon prevention for new buildings, as well as the promotion of actions for the population aimed at reducing exposure to radon.

RPAs are most commonly estimated using indoor radon data, but also geogenic data (i.e. uranium concentration in the ground, terrestrial gamma dose rate, geological units, soil units and others) could be used together with indoor radon data to benefit of the synergic information. These predictors or proxy quantities are physically and statistically related to indoor radon quantities and are at the base of the concept of GRP [46]. Therefore, also atmospheric radon concentration and radon fluxes could be considered geogenic variables useful to estimate the GRP and to identify RPAs.

The first activity consisted in an extensive literature research of outdoor radon and radon flux, which resulted in two published papers [47]. The literature review showed positive correlations of outdoor radon as well of radon flux with other radon quantities (radon in soil gas, indoor radon concentration), which indicates a possible relevance for the identification of RPA. European radon flux maps already have an excellent spatial and temporal resolution, could serve as input parameter for the delineation of RPA. Moreover, radon flux has shown to be an important input parameter for the prediction of averaged.

As a result, it was found that long-term averages of outdoor radon show strong correlations with indoor radon concentrations and other radon parameters. Taking into account the very limited amount of available outdoor radon observations for the analysis, the significance of outdoor radon for radon risk prediction is likely to increase with additional observations. After collecting data it was compared to outdoor radon and radon flux with 28 other parameters used for radon risk prediction such as geological information, physical and chemical soil properties and weather data. The gridded indoor radon concentrations of the European Atlas of Natural Radiation [48] was used as proxy for the radon potential of an area and as target variable in a machine learning workflow.

Following an approach outlined by [49] and using a random forest model for prediction on a data set covering the area of Belgium and Germany was used. A sufficient number of outdoor radon measurements are already available in these two countries. Repeatedly 500000 random forest models with different input features and evaluated the model performance were built. The German data set was used to train the model and as well to evaluate the model performance in a five-fold cross validation. The Belgian data was only used for performance evaluation. This reproduces one of the core ideas of an European radon potential map, where a model developed in one country can also be used in another country to predict the radon potential.

To evaluate the model performance the mean square error of predictions and actual target values was used. The models that performed best on the Belgian test data and German validation data set were selected and optimized. These two models only share one input feature (soil moisture) but otherwise use different input features. The Belgian model uses the radon flux for prediction. The impact of the input features shows high variabilities, where for the German model the performance gain of a single input feature is in the range of a few percent, whereas for the Belgian model a single input feature (the number of coarse fragments in the top soil) more than doubles the model performance. The choice of input features to be used as predictors for the radon potential strongly depends on the setting and area. A predictor proven to work in one area might not necessarily work in another, and there is still a potential for input features not selected by our models to work in other areas. The statistics on the 100 best performing models also reviled that both radon flux and outdoor radon frequently occur in these best models. There is a potential for these two parameters in radon risk prediction. Still other parameters might be more important, but radon flux and outdoor radon contribute valuable information. An increase of measurements and observations of radon flux and outdoor radon is highly appreciated, which is likely to increase the impact of these quantities for radon risk mapping. Compared to other quantities the observation density of radon flux and outdoor radon is very low, but the consortium expects also based on the results of the traceRadon project an increase of measurements in the near future.

With the established metrology for reliable measurements of outdoor radon and radon flux within the traceRadon project, long-term as well as data with higher spatial resolution will be available hopefully in the mid-term future. Standard measurement protocols for radon flux and outdoor radon should be developed across Europe to increase comparability of different measurement campaigns and the usability for RPA prediction.

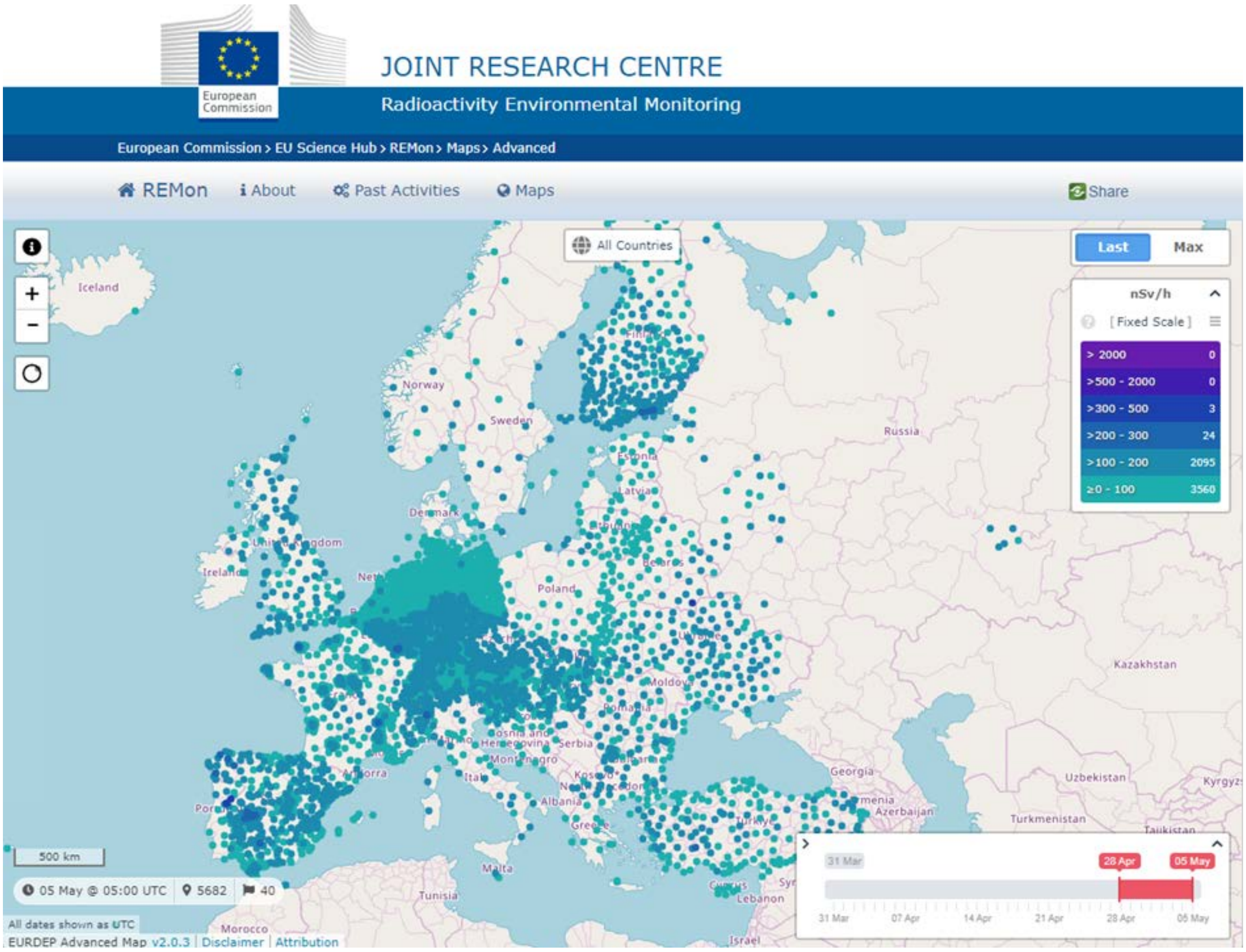

Figure 4: Screenshot of the EURDEP system displaying the location of gamma dose rate sensors at European countries between 28 April and 5 May 2022. Reproduced from remap.jrc.ec.europa.eu, Advanced Map. CC BY 4.0. Accessed 5 May 2022.

The second application is related to ambient gamma dose measurements for environmental radioactivity monitoring. These kinds of measurements are widely used in nuclear or radiological emergency preparedness and response systems [50]. And in this kind of monitoring, it is fundamental to avoid false positives caused by natural phenomena and to be as precise as possible in the quantification of components of environmental radiation. The EURDEP (European Radiological Data Exchange Platform) could be considered an example of a network susceptible to such problems (see Fig. 4). It is a system for the exchange of radiological monitoring data from automatic surveillance systems in 39 countries in almost real time [50], mostly by non-spectrometric detectors such as Geiger-Muller or proportional counters which cannot distinguish between radiation sources. To carry out this delicate task, the EURDEP has been developed and improved over the past 30 years. Assuming no radiological/nuclear events occur, these measurements, in the form of dose rate (terrestrial and atmospheric gamma and cosmic muons), essentially reflect the natural background gamma/muon radiation from approximately 5500 fixed sensors, see Fig. 5.

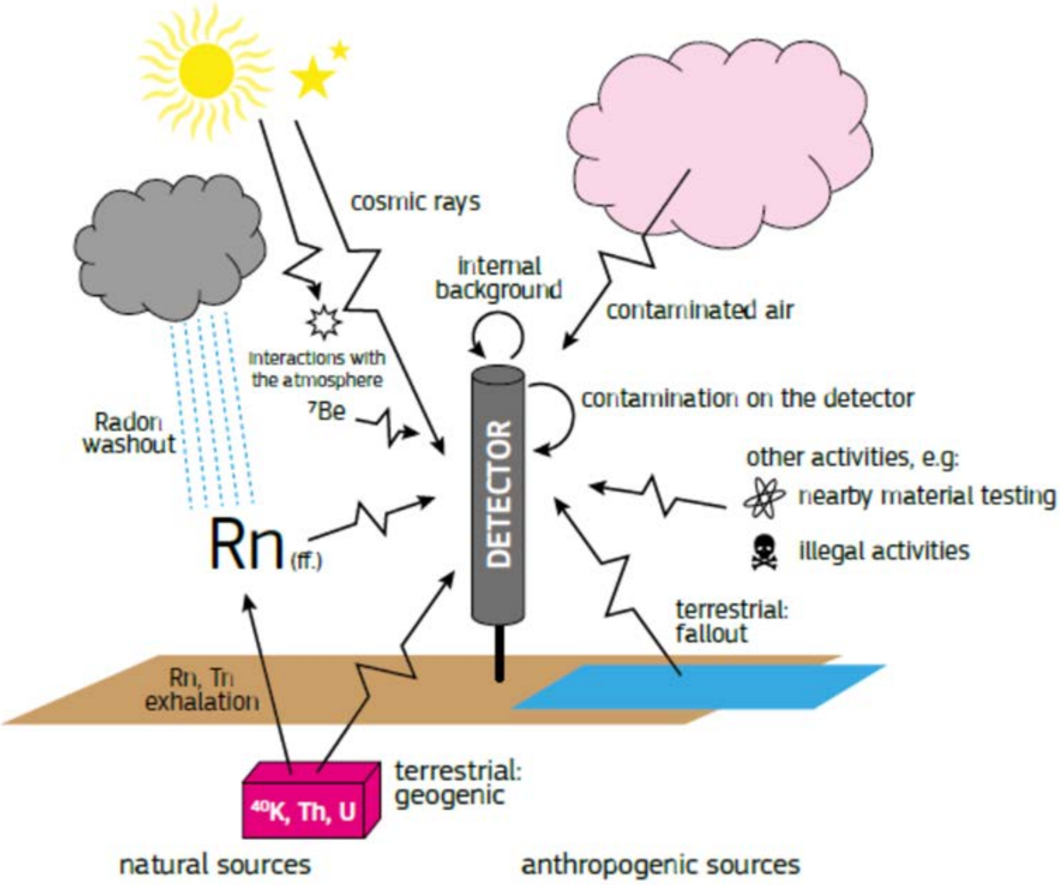

Figure 5: Graphic summary of the contributions to the dose rate recorded by a detector. Rn – radon (222Rn), Tn – thoron (220Rn). Source European Atlas of Natural Radiation (EC 2019).

The EURDEP provides current and continuous information available on https://remon.jrc.ec.europa.eu/. However, so far, the system does not have a primary alerting role, and hence, cannot automatically be taken as an indication of increased levels of radioactivity without prior consultation with the data providers. In fact, peaks in ambient dose rate (ADR) due to natural temporal variation can create false positive responses in a network such as EURDEP. The most important examples are the so-called radon wash-out peaks due to the deposition on the ground of solid radon progenies by rain or snow lead to an increase in the observed GDR [51].

In this context atmospheric radon and radon flux data together with different methods (i.e. artificial intelligence), could help to quantify, simulate, and automatically remove these natural peaks. This will help both to prevent false alarms and to improve the detection in case of radiological/nuclear accidents by enabling the reduction of the detection threshold. In order to better understand radon wash-out peaks and try to prevent false alarms in gamma dose rate monitoring systems such as EURDEP, an inter-comparison exercise was organized in the framework of the traceRadon project. It represents the first inter-comparison of different automatic methods to identify and to classify ambient dose rate peaks, hence also radon wash-out peaks. It has been organized in collaboration with researchers from EURADOS (European Radiation Dosimetry Group, https://eurados.sckcen.be/). Six participants coming from different institutions over Europe agreed on taking part to the exercise. The exercise was carried out in two rounds. In the 1st round, a 3-month time series of ambient dose equivalent rate $H^*(10)$ data with hourly resolution were provided to the six participants which had to design and develop method and criteria to reach the intercomparison exercise goal. In the 2nd round rain time series data were also provided to the participants. Each participant could provide peak occurrence dates (start-end) and their classification (natural, artificial, etc.). Main conclusions from the exercise are:

- sensitivities for peak identification increased for each method between 1st and 2nd round;
- the percentage of peaks identified during the 2nd round ranged from 80% to 100% (except participant 6 with 50%);
- radon wash-out peaks were identified by all the participants during the 2nd round; the average duration of radon wash-out peaks ranged between 3 and 9 h;
- nocturnal radon accumulation peaks identification and classification highly improved during the 2nd round; the average duration of nocturnal radon accumulation peaks was around 10 h;
- suspect-anomaly peaks were identified by most of participants at the 1st round and the classification highly improved during the 2nd round. The average duration of suspect-anomaly peaks was lower than 5h;
- spectrometric data are necessary to know the origin of the peaks (natural or artificial);
- rain information is fundamental for a good classification of the peaks (i.e., most of the nocturnal radon accumulation peaks were classified as rain wash-out peaks during the 1st round, whereas rain peaks were classified as suspect-anomaly peaks).

In conclusion, the objective was fully met: The new maps are available for the use in radiation protection, the new approach for RPA is implemented and new algorithms to prevent false alarms in EURDEP are available.

All partners (JRC, PTB, VINS, AGES, CLOR, INESC TEC, LUND, UC, UoB, UPC) collaborated in the generation of these results. Moreover, 19ENV01 traceRadon has been proven to be a good platform to foster collaborations among different groups. By building on the current experience and promoting community work it is expected that additional analysis can be efficiently conducted to improve the currently available methods. Since the identification of the peaks is particularly challenging, these results could constitute a benchmark for future developments.

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# 5 Impact

The project has created a website at http://traceradon-empir.eu/ and a traceRadon newsletter in order to promote itself to end users. A project Twitter account was created https://twitter.com/traceradon as well as a notice board on ResearchGate until ResearchGate removed the possibility to have project webpages. Further to this, the project has been presented 71 times at conferences and events such as the Atmospheric Composition & Chemistry Observations & Modelling Conference, the Romanian Society for Radiological Protection, the ICOS MSA (Monitoring Station Assembly) Atmosphere Meeting, the Sensor and Measurement Science International, the EGU General Assembly 2021, 2022, 2023 in the session of geoscience applications of environmental radioactivity, the 20th international metrology congress CIM 2021, the 15th International Workshop on the Geological Aspects of Radon Risk Mapping, the Final Conference LIFE-Respire, 6th European IRPA Congress, ICRM-LLRMT 2022, ICRM 2023, EURADOS WG3-S3 2021, 2022, 2023, ICOS Science Conference 2022, the IAEA: Second International Conference on Applications of Radiation Science and Technology (ICARST 2022), the Conference on Climate Change Impact on Radon and Human Health Dose Assessment, Yukon University, the IMEKO Conference of TC8, TC11, TC24 and many more. The project presented results on all continents, in different communities and receives even after its end still applications to join.

Moreover, several invited talks have been given by the project partners to stakeholders such as the Radiation protection platform (Austria), the Sciences du Climat et de l'Environnement (LSCE, France), the European Radon Week with the 9th ERA Workshop on International Collaborations on Radon, the 2nd HERCA Workshop on National Radon Action Plans, and the European Atlas of Natural Radiation (EANR). These talks have been the result of dissemination of the project, including four newsletters and six articles in the popular press. So far interest in the project has comes from a broad range of different sectors: legislation, health and climate protection, physics and geology as well as voluntary organisations.

A highlight on the political level was the Delegation trip of the coordinator with Prime Minister of Lower Saxony Stephan Weil to Oslo/Norway from May 21 to 24, 2023 together with Olaf Lies, Minister for Economic Affairs, Transport, Building and Digitalization and to Tallinn/Estonia, May 24-26, 2023, which gave the opportunity to address the metrology needs for the future to combat climate change.

*Impact on industrial and other user communities*

European climate observation groups and radiological protection groups both benefited from this project i.e. (i) climate related Atmospheric Monitoring Network stations (AMNS) (e.g. ICOS), and (ii) the European Radiological Data Exchange Platform (EURDEP) and the EANR. By improving the traceability of low-level radon and radon flux measurements this project supported collaboration between these currently independent groups. Such interdisciplinary collaboration provided new insight and understanding on the links between geology, the atmosphere, and anthropogenic activity and their combined impact.

Accurate knowledge of environmental outdoor radon activity concentrations and radon flux is key for improving Greenhouse Gas (GHG) flux estimates for climate observation and radiological protection. Current climate related AMNS were established for measurements of GHGs in order to support interpretation of the ATM (Atmospheric Transport Model) and to better understand GHG levels using long-term observations. Atmospheric radon measurements are carried out at such AMNS and therefore, this project supported European AMNS in performing atmospheric radon and radon flux measurements for a variety of radon tracer applications. The project did this through its development of new low activity Rn-222 emanation sources, a reference instrument for atmospheric radon measurements and a traceability chain for low radon activity concentration measurements (from 1 Bq $m^{-3}$ to 100 Bq $m^{-3}$). All of which supported the comparability of real time atmospheric radon activity concentration data between different measurement sites and over time provide these radon measurements with the required traceability to the SI.

To support its engagement with industry and other user communities the project has set-up a Stakeholder Committee which currently has 20 members and includes high impact, multi- national stakeholders such as: the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), the World Meteorological Organisation (WMO), the International Commission on Radiological Protection (ICRP), the International Committee for Radionuclide Metrology (ICRM), the European Radon Association (ERA), the European Radiation Dosimetry Group (EURADOS) and EURAMET’s Technical Committee for Ionising Radiation (TC-IR). Further to this, the stakeholder committee also includes standardisation bodies such as the Commission Electrotechnique Internationale (IEC) and the International Organisation for Standardisation (ISO) as well as representative organisations from individual nations e.g., ANTSO, Australia’s Climate Science Centre, Oceans & Atmosphere (CSIRO), Germany’s Federal Office For Radiation Protection (Bundesamt für Strahlenschutz), Germany’s Meteorological Service (Deutscher Wetterdienst), Spain’s Centre for Energy, Environmental and Technological Research (CIEMAT), Environment and Climate Change Canada, Ireland's Environmental Protection Agency Office of Radiation Protection and Environmental Monitoring, the UK's Society for Radiological Protection (SRP), the UK's Met Office, the National Metrology Institute of South Africa (NMISA), Japan's National Institutes for Quantum and Radiological Science and Technology (QST), Italy's Politecnico di Milano - Department of Energy, Italy's National Research Council/ Biometeorology lab (IBIMET-CNR), Romania's National Commission for Nuclear Activities Control (CNCAN) and the University of Novi Sad (Serbia).

Further to this the project has received interest from stakeholders in the uptake of its results. These include the Federal Office for Radiation Protection (Bundesamt für Strahlenschutz, Germany), who is interested in the results of the source development for its calibration services. The company Radonova Laboratories AB (a leader in radon measurement based in Sweden) is interested in the project's research activities and capacity building. A prototype detector developed in the scope of the project is on its way to be commercialized by Radonova, too.

Finally, the European Radon Association (ERA) is interested in the project's development of new method to identify RPAs.

*Impact on the metrology and scientific communities*

The project's data on outdoor radon activity concentration and radon flux measurements can be used to provide key information on atmospheric radon activity concentrations; one of the greatest natural radiological risks. The project's data was made available online for scientists, policy and decision makers and end users. The project's developments in techniques for measuring low-level environmental radon activity concentration and radon fluxes is and will be useful for the metrological community working in this field, for regulatory authorities, civil protection or official measuring bodies, and for manufacturers of radon monitors or dosimeters. In addition, the project has significantly advanced radon flux metrology. It did this by providing a calibration infrastructure, including a radon exhalation reference system "exhalation bed" and a transfer standard. The project used this capability to harmonise existing radon flux instruments/methods using field-based comparisons. Radon flux measurements carried out over Europe during the project validated existing European radon flux models and inventories in order to obtain online real-time European radon maps. These radon and radon flux maps are now available for atmospheric studies and for radiological protection such as the identification of RPAs.The project has provided training to the metrology and scientific communities at sixteen events such as:

Workshops (8): Two general scientific workshops on traceRadon, New Procedures for Radon Monitoring, New Procedures, guidelines an methodologies for radon instruments calibration and measurements, Gap workshop oon radiation protection metrology, traceRadon as a tool for the national GHG strategy for Hungary, Precise and traceable Radon activity concentration measurements, Precise and traceable measurement of Rn flux and the application of the Radon Tracer Method, Strategy on making available the radon flux campaign data

Training courses (4): New procedures, guidelines and methodologies for radon instrument calibration and measurements, Radioactivity and radiation: New methods for climate observation and climate modelling. Details in opperation of radon and radon flux monitors, Radon measurements in the Arctic: the challenges, technology and research benefits

Internal training for members and collaborators (3): Operation and calibration of the ANSTO monitors and the new prototype the ANSTO 200L, Installation of Radon Flux Campaign Equipment: Technical Training, Data Review of the Field Campaign

Further to this the project was collaborating with organisations in the scientific community including the EMN Climate and Ocean Observation, ERA, EURADOS, the UK's Metrological Office, the Universität Heidelberg, Germany, the University of Novi Sad, Serbia, the Politecnico di Milano, Italy, the University of Codoba, Spain, the Universität Siegen, Germany, Institut de radioprotection et de sûreté nucléaire (IRSN, responsible for performing radiological monitoring of the environment throughout France), The Regional Environmental Protection Agency (Agenzia regionale per la protezione ambientale, ARPA Valle d'Aosta and ARPA Piemonte) Italy, Radonova, Sweden, the LIFE-Respire, retired expert Peter Bossew and Ulrich Stöhlker, the University of Groningen and ANSTO, Australia.

*Impact on relevant standards*

The project has provided input to HERCA (Heads of the European Radiological Protection Competent Authorities), the Federal Ministry for the Environment, Nature Conservation and Nuclear Safety (BMU), DKE GK 851 Activity measuring devices for radiation protection, ISO/TC 85 Nuclear energy, nuclear technologies, and radiological protection, BIPM and CIPM CCRI (I) (x- and gamma rays, charged particles),CIPM CCRI (Measurement of radionuclides), CIPM CCRI (Consultative Committee for Ionising Radiation) in particular the CCRI Strategy 2018-2028, EURAMET TC-IR (Ionising Radiation), EURADOS WG3 Environmental dosimetry, EURATOM, the Professional Association for Radiation Protection Environmental Monitoring Working Group and the European Radioecology Alliance Topical Working Group Atmospheric radionuclides in transfer processes.

In addition, the project provided input to: IEC/TC 45 Nuclear Instrumentation SC45B Radiation protection instruments, WG9 Detectors and systems, ISO/TC 142 Cleaning equipment for air and other gases ISO/TC146 Air quality and related activities and ICRM (Gamma-Ray Spectrometry WG, Alpha-Particle Spectrometry WG and Low Level Measurement Techniques WG).

*Longer-term economic, social and environmental impacts*

Climate change and radiological protection both affect humankind and the environment, worldwide. For the planet to combat both climate change and radiation exposure, measurements must be supported by reliable metrology. By addressing a topic (i.e. the measurement of low levels of radon in the environment) that supports both climate observation and global radiological protection, this project simultaneously supports the long-term economic, social and environmental work of ICOS, the Integrated Pollution Prevention and Control (IPPC) Directive 2008/1/EC, the IAEA, Analytical Laboratories for the Measurement of Environmental Radioactivity (ALMERA) and WHO.

The project’s data on low level measurement of radon in the environment improved ATMs and their ability to estimate GHGs fluxes which in turn supports the EU Emissions Trading System (EU ETS). The EU ETS is a cornerstone of the EU's long-term policy to tackle climate change through a cost-effective reduction of emissions of carbon dioxide ($CO_2$) and other GHG in the power, aviation and industrial sectors. The projects results will thus support Europe in its movement towards a competitive low carbon economy.  At the same time, the project has provided the EC (through project partner the JRC) with access to reliable data of outdoor radon activity concentrations, which can be used in combination with soil exhalation flux measurements, for dynamic mapping of radon in the environment. By supporting the provision of accurate knowledge of RPA this project supported European radiation protection measures and thus in longer-term help to lower radiation protection costs. The EC JRC has taken the already taken the approach to their information page in the Digital Atlas of Natural Radiation, subsection radon flux, Monthly maps display the radon flux from the earth for atmospheric tracer transport and radon protection analysis on a 0.05° x 0.05° grid.

# 6 List of publications

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**List of publications**

1. Mertes, F et. al.: D3.3 Approximate sequential Bayesian filtering to estimate Rn-222 emanation from Ra-226 sources from spectra, https://doi.org/10.5162/SMSI2021/D3.3
2. Röttger, A. et al: New metrology for radon at the environmental level *2021* Meas. Sci. Technol. *32,* 124008, https://doi.org/10.1088/1361-6501/ac298d
3. Radulescu, I et al.: Inter-comparison of commercial continuous radon monitors responses, Nuclear Instruments and Methods in Physics Research Section A, Volume 1021, 2022, 165927, https://doi.org/10.1016/j.nima.2021.165927
4. Mertes, F. et. al.: Ion implantation of $^{226}$Ra for a primary $^{222}$Rn emanation standard, Applied Radiation and Isotopes, Volume 181, March 2022, 110093, https://doi.org/10.1016/j.apradiso.2021.110093
5. Čeliković, I. et. al.: Outdoor Radon as a Tool to Estimate Radon Priority Areas - A Literature Overview, Int. J. Environ. Res. Public Health 2022, 19, 662, https://doi.org/10.3390/ijerph19020662
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